US011604293B2

(12) United States Patent
Freeman

(10) **Patent No.: US 11,604,293 B2**

(45) **Date of Patent: Mar. 14, 2023**

(54) **BORON TRIFLUORIDE AS A QUENCH GAS FOR NEUTRON PROPORTIONAL COUNTERS**

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(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 33 days.

(21) Appl. No.: **17/401,988**

(22) Filed: **Aug. 13, 2021**

(65) **Prior Publication Data**

US 2023/0045923 A1 Feb. 16, 2023

(51) **Int. Cl.**
G01T 3/00 (2006.01)

(52) **U.S. Cl.**
CPC ***G01T 3/008*** (2013.01)

(58) **Field of Classification Search**
CPC G01T 3/00; G01T 3/008; G01T 1/185; G01T 1/18; G01T 1/205; H01J 47/12; H01J 47/06; H01J 47/02; H01J 47/08; H01J 47/005
See application file for complete search history.

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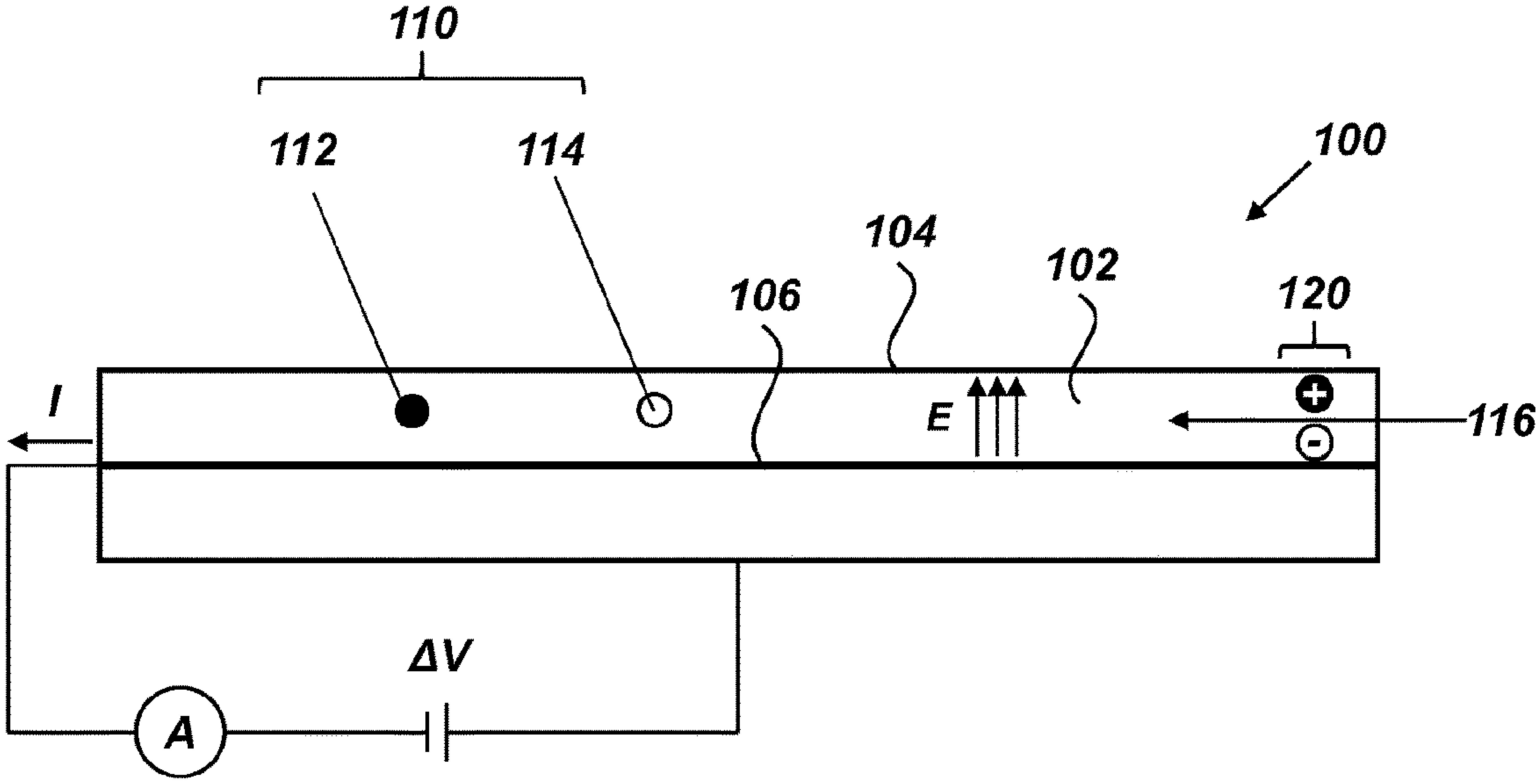
(57) **ABSTRACT**

A neutron proportional counter is provided. The proportional counter can include a chamber and a gas mixture. The chamber includes an anode and a cathode. The gas mixture is contained within the chamber and includes at least one neutron sensitive fill gas and a quench gas including BF_3. In certain embodiments, the neutron sensitive fill gas can be configured for detection of thermal neutrons (e.g., He-3), fast neutrons (e.g., He-4, H_2), or both (e.g., UF_6).

18 Claims, 2 Drawing Sheets



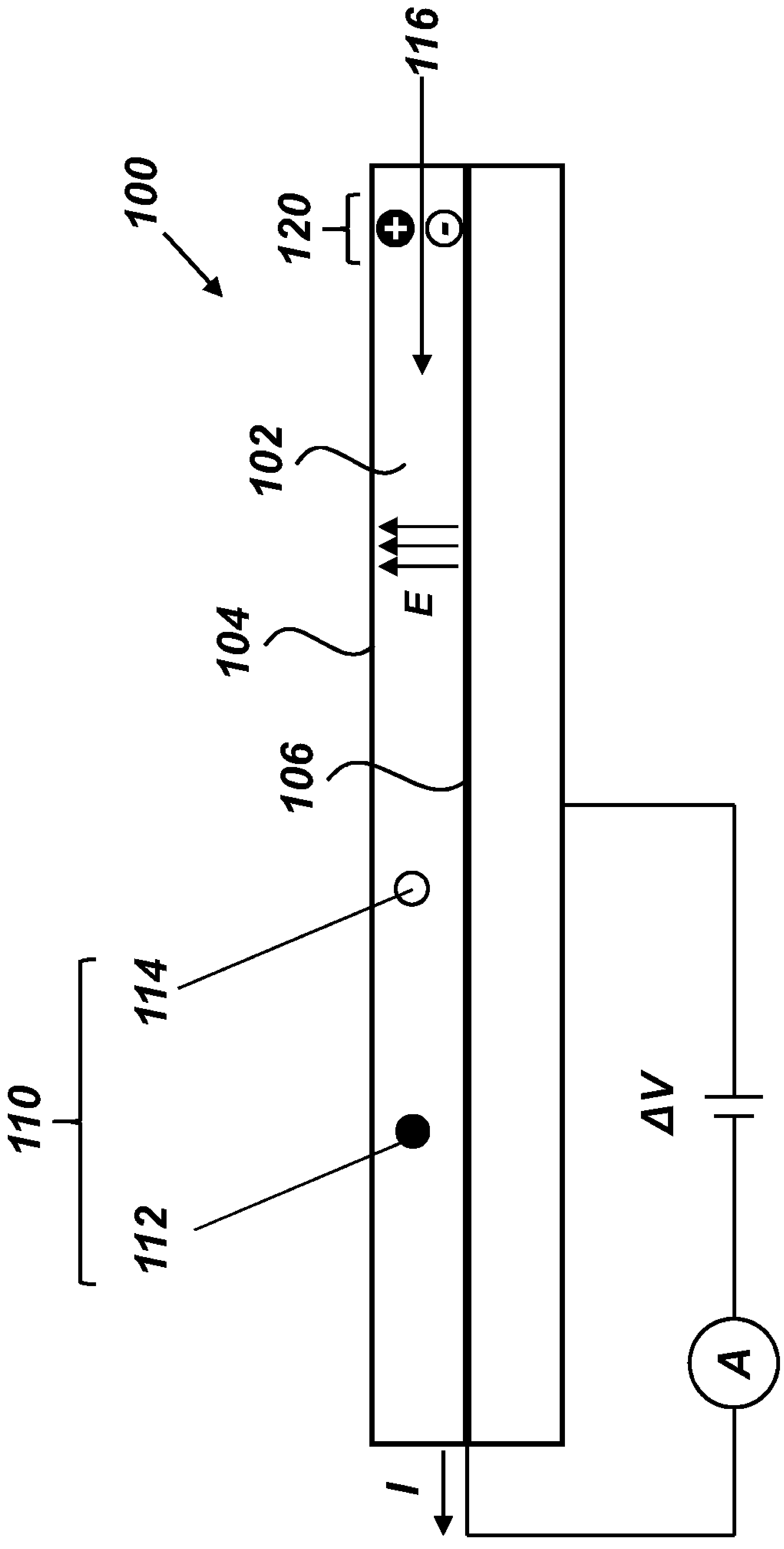


FIG. 1

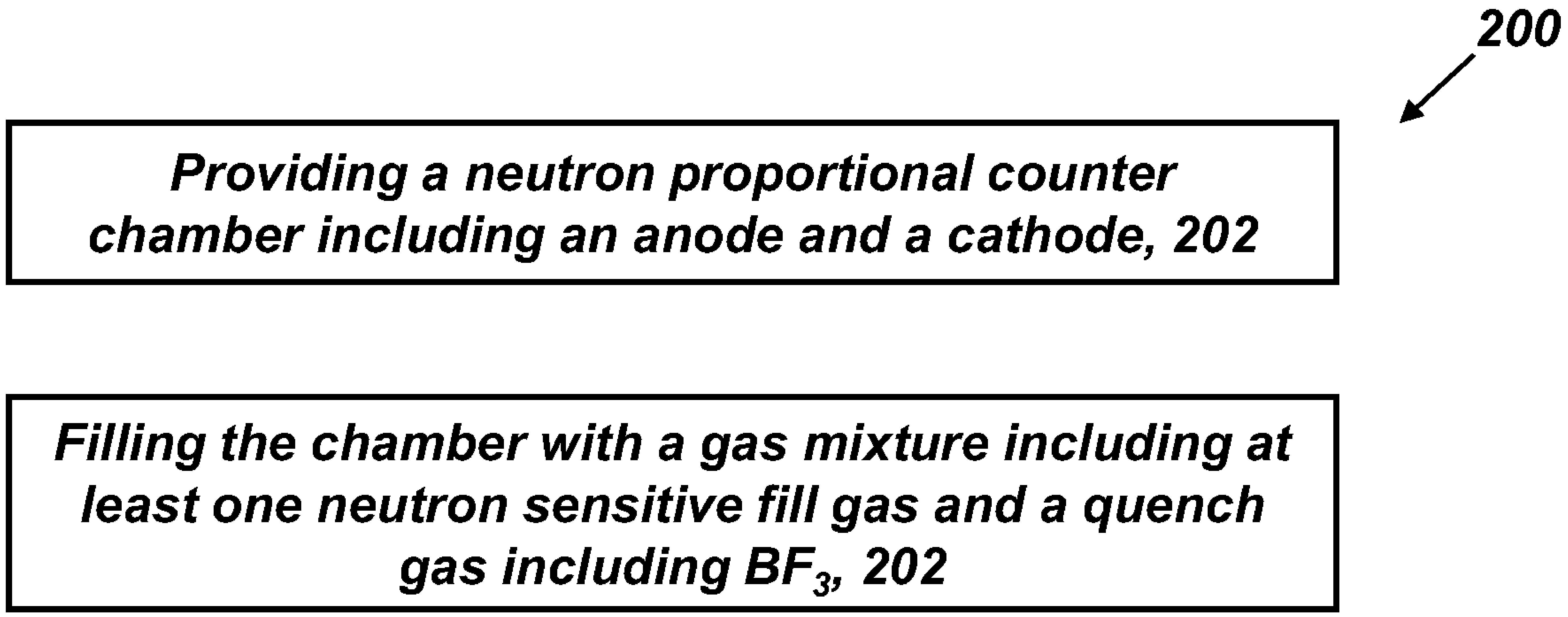

200
Providing a neutron proportional counter chamber including an anode and a cathode, 202
Filling the chamber with a gas mixture including at least one neutron sensitive fill gas and a quench gas including BF_3, 202

FIG. 2

BORON TRIFLUORIDE AS A QUENCH GAS FOR NEUTRON PROPORTIONAL COUNTERS

BACKGROUND

Ionizing radiation is a type of energy released by atoms in the form of electromagnetic waves (e.g., gamma rays, X-rays) or particles (e.g., neutrons, alpha particles, beta particles) during disintegration of atomic nuclei. Sources of ionizing radiation are found in nature (e.g., radioactive materials in soil, water, air, cosmic rays, etc.), as well as man-made sources (e.g., nuclear power generation, medical radiation, etc.)

SUMMARY

Gaseous ionization detector are radiation detection instruments used in particle physics to detect the presence of ionizing radiation particles, as well as in radiation protection to measure ionizing radiation. Proportional counters are a type of gaseous ionization detector and are commonly used when discrimination between types of radiation (e.g., alpha vs. beta particles) is desired, as well as circumstances where pre-electrical signal amplification, improved signal-to-noise ratio, and/or enhanced noise discrimination is desired.

Embodiments of the disclosure provide an improved proportional counter and corresponding methods. As discussed in detail below, the proportional counter includes a novel gas mixture that provides the improved detection sensitivity (e.g., sensitivity to thermal neutrons), as compared to proportional counters employing conventional gas mixtures.

In an embodiment, a neutron proportional counter is provided. The proportional counter can include a chamber and a gas mixture. The chamber can include an anode and a cathode. The gas mixture can be contained within the chamber and the gas mixture can include at least one neutron sensitive fill gas, and a quench gas including BF_3.

In another embodiment, the fill gas can have a thermal neutron absorption cross-section within the range from about 100 to 5600 barns.

In another embodiment, the fill gas can have a fast neutron total cross-section within the range from about 1 to 8 barns.

In another embodiment, the fill gas can be He-3. He-3 can be present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 1.5 Psia to about 150 Psia.

In another embodiment, the fill gas can be at least one of He-4, H_2, or UF_6.

In another embodiment, the gas mixture can further include at least one stopping gas configured to reduce the mean free path of primary ions within the gas mixture. In another embodiment, the stopping gas can be at least one of Ar, Kr, or Xe.

In another embodiment, BF_3 can be present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 0.002 Psia to about 3.9 Psia.

In an embodiment, a method of preparing neutron proportional counter is provided. The method can include providing a neutron proportional counter. The proportional counter can include a chamber comprising an anode and a cathode, and filling the chamber with a gas mixture. The gas mixture can include at least one neutron sensitive fill gas and a quench gas including BF_3.

In another embodiment, the fill gas can have a thermal neutron absorption cross-section within the range from about 100 to 5600 barns.

In another embodiment, the fill gas can have a fast neutron total cross-section within the range from about 1 to 8 barns

In another embodiment, the fill gas can be He-3. He-3 can be present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 1.5 Psia to about 150 Psia.

In another embodiment, the fill gas can be at least one of He-4, H_2, or UF_6.

In another embodiment, the gas mixture can further include at least one stopping gas configured to reduce the mean free path of primary ions within the gas mixture.

In another embodiment, the stopping gas can be at least one of Ar, Kr, or Xe.

In another embodiment, BF_3 can be present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 0.002 Psia to about 3.9 Psia.

DESCRIPTION OF DRAWINGS

These and other features will be more readily understood from the following detailed description taken in conjunction with the accompanying drawings, in which:

FIG. **1** is a diagram illustrating one exemplary embodiment of proportional counter including a gas mixture including an He-3 fill gas and a BF_3 quench gas; and

FIG. **2** is a flow diagram illustrating one exemplary embodiment of a method of preparing the proportional counter of FIG. **1**.

It is noted that the drawings are not necessarily to scale. The drawings are intended to depict only typical aspects of the subject matter disclosed herein, and therefore should not be considered as limiting the scope of the disclosure.

DETAILED DESCRIPTION

A proportional counter is provided for detection of radiation (e.g., neutrons) that includes a novel gas mixture. As discussed in detail below, the proportional counter can include a gas mixture including a neutron sensitive fill gas and a quench gas including BF_3. In one aspect, the gas mixture provides improved sensitivity for detection of neutrons, as compared to gas mixtures without BF_3. In another aspect, decomposition products of the BF_3 quench gas are capable of recombining, providing the quench gas with effectively indefinite life.

FIG. **1** illustrates one exemplary embodiment of a proportional counter **100**. As shown, the proportional counter **100** includes a chamber **102** having two electrodes (e.g., a cathode **104** and an anode **106**) separated from one another. The chamber **102** can further contains, or be filled to contain, a gas mixture **110** under pressure. That is, the chamber **102** can include at least one port (not shown) configured to couple to a gas source for receiving the gas mixture **110** or components thereof. The chamber **102** can further include one or more seals (not shown) configured ensure that the chamber **102** is substantially fluid tight, inhibiting egress of the pressurized gas mixture **110**.

In an embodiment the gas mixture **110** can include a fill gas **112** (e.g., a neutron sensitive gas) and a quench gas **114**. The fill gas **112** can be configured to interact with, or be ionized by, incident radiation **116**. The quench gas **114** can be configured to terminate pulse discharge.

In use, a voltage ΔV is applied between the electrodes **104**, **106** to generate an electric field E therebetween.

Incident radiation **116** (e.g., a neutron) entering the proportional counter **100** can collide with a molecule of the fill gas **112** and ionizes it to produce an electron − (primary electron) and a positively charged atom or molecule +, collectively ion pair **120**, also referred to herein as primary ions. The voltage ΔV is sufficient so that conditions within the chamber **102** correspond to the proportional region of the counter **100**. In one aspect, the electric field strength is sufficiently high to prevent recombination of ion pairs, causing positive ions to drift towards the cathode and electrons towards the anode. In the vicinity of the anode, the field strength is also large enough to accelerate the primary electrons, causing ionization of additional atoms of the fill gas **112**, generating further ion pairs (including secondary electrons). The electrons − collected at the anode **106** form the output (e.g., ion current I) of the proportional detector **100** and can be measured by an ammeter A or other current measuring device. Beneficially, in the proportional region, each ionizing particle produces only one avalanche. Thus, proportionality is provided between the number of events (ionizing particles) and the total ion current I. Additionally, the charge amplification provided by the avalanche improves the signal to noise ratio of the proportional detector **100** and reduces the amount of subsequent signal amplification required.

Embodiments of the present disclosure propose a gas mixture **110** that includes a neutron sensitive gas as the fill gas **112** (e.g., helium-3) and BF_3 as the quench gas **114**. As discussed in detail below, this combination is counter to conventional wisdom as understood by one skilled in the art and provides advantages over use of organic gases as the quench gas **114**.

Some early proportional detectors have employed boron tri-fluoride BF_3 as a fill gas. Boron-10 exhibits excellent thermal neutron capture cross-section, making is suitable for use to measure thermal neutron flux. In thermal neutron boron reaction:

$$^{10}B+^{1}n\rightarrow[^{11}B]\rightarrow^{4}He+^{7}Li+2.8\text{ MeV}$$

boron-10 (^{10}B) absorbs a neutron (^{1}n) to yield boron-11 (^{11}B). Boron-11 subsequently decays into reaction products helium-4 (^{4}He) and lithium-7 (^{7}Li) and 2.8 MeV (gamma rays). The short range of the reaction products means that the total energy can be collected in a relatively short distance. This allows for discrimination of lower energy gamma rays from the thermal neutron signals in a relatively small package.

Use of BF_3 as the fill gas in proportional detectors has largely been supplanted by helium-3, however. In one aspect, as compared to BF_3, helium-3 exhibits a significantly higher capture cross-section. In another aspect, significant care can be required when handling BF_3, as it is hazardous (e.g., toxic by inhalation) and corrosive, as it can form highly corrosive hydrofluoric acid when dissolved in water. Thus, for many detection tasks, neutron detection is more easily performed using helium-3.

The quench gases used in combination with helium-3 are typically organic gases (e.g., carbon dioxide CO_2, methane CH_4, carbon tetrafluoride CF_4). In general, quenching action describes when the energy from the pulse is dissipated by the quench gas. This dissipation occurs when the molecules of the quench gas use the energy of the incident radiation to break apart (or dissociate) into their separate elements, rather than ionize or re-release another photon (as do other gases in the gas mixture).

While organic gases are suitable quenchers for many applications of the proportional counter, they can exhibit some drawbacks. In one aspect, these organic gases are consumed over the life of the detector due to the nature of their dissociation as they quench. That is, once these molecules have split, they are not capable of reforming. Furthermore, in some cases, the split molecules can result in deposition of carbon on internal components of the proportional counter and degrade its performance.

In another aspect, it will be appreciated that these organic gases function solely as quenchers. That is, they represent a portion of the total gas in the detector that is not sensitive to thermal neutrons. For detector designs that are limited by maximum pressure of the gas mixture **110**, the amount of quench gas **114** must be carefully balanced by the sensitivity required for the desired detection application.

Accordingly, there exists a need for gas mixtures including different quench gases exhibiting improved performance as compared to the above-discussed organic gases. Embodiments of the present disclosure propose a gas mixture **110** that includes a neutron sensitive gas as the fill gas **112** (e.g., helium-3) and BF_3 as the quench gas **114**. In certain embodiments, the only quench gas is BF_3. As further discussed below, use of BF_3 as the quench gas is unconventional, given acknowledged drawbacks of BF_3 as a fill gas. However, BF_3 offers compelling advantages when used in as the quench gas **114** in combination with the fill gas **112**.

In general, BF_3 is an electronegative gas. Too much BF_3 can drive the voltage required for operation too high to be useable (e.g., out of the proportional range). That is, because of the quenching functionality of BF_3, the more that is added, the higher the voltage ΔV needed to operate as a detector for alpha particles (^{4}He). For this reason, the pressure of BF_3 in use is generally limited to about 1 atmosphere when used unmixed in a proportional counter.

In another aspect, too much BF_3 can slow the response of the counter. As an example, the speed with which the proportional counter collects all the charge from a given event is determined by the drift velocity of electrons in the fill gas, and the drift velocity changes for a given gas mixture. As BF_3 is not a gas that gives rise to relatively fast drift velocity, too much of it in the gas mixture can slow charge collection, and therefore limit the speed with which the proportional counter can resolve individual events

In a further aspect, too much BF_3 can potentially alter the shape or number of pulses from a single event. Without being bound by theory, it is believed that the combination of low drift velocity and electronegativity of BF_3 results in failure to capture the full energy of the pulse in the same time period as the rest of the pulse.

In an additional aspect, as discussed above, helium-3 has a much higher cross-section for thermal neutron capture as compared to BF_3 and can be filled to higher pressures. Thus, in instances where the gamma field is low enough to permit use of helium-3, BF_3 provides no advantages if substituted for helium-3 as the fill gas to absorb thermal neutrons.

However, use of BF_3 as the quench gas **114** in the gas mixture **110**, as compared to traditional organic gases, has a variety of advantages, discussed in detail below.

In one aspect, because BF_3 is a halogen quench agent, its decomposition products are capable of recombining. As a result, a negligible amount of BF_3 is consumed when quenching, giving it an effectively indefinite life, even in high-flux environments.

In another aspect, because BF_3 is at least thermal neutron sensitive, its use as the quench gas **114** serves to improve at least the total thermal neutron sensitivity of the proportional detector **100**.

In a further aspect, as boron-10 deposits more energy per neutron interaction than helium-3, the presence of BF_3 can help to mitigate the sensitivity loss experienced by helium-3 proportional counters when employed in moderate gamma fields (e.g., about 10 to about 1000 R/hr). A t these gamma radiation levels, the pulse height discrimination level should be raised to remove gamma interference. Accordingly, some fraction of helium-3 pulses can be discriminated out, but higher energy pulses from boron-10 would not be.

BF_3 can be present within the gas mixtures **110** in an amount sufficient to provide a partial pressure within the range from about 0.002 Psia to about 3.9 Psia. The lower bound represents the minimum partial pressure of BF_3 sufficient for quenching. This pressure corresponds to the pressures typically used for halogen quenching in a Geiger-Müller tube. The upper bound represents the maximum partial pressure of BF_3 before BF_3 effectively dominates over the fill gas **112**. That is, before the operational characteristics of the gas mixture **110** approaches that of pure BF_3. It can be appreciated that these partial pressures are provide as examples and that the minimum/maximum partial pressure of BF_3 can adopt other values in view of detector size, detector construction, desired sensitivity, and maximum pressure.

Embodiments of the gas mixture can employ a variety of neutron sensitive gases as the fill gas **112** in combination with BF_3 as the quench gas **114**. In certain embodiments, the neutron sensitive gas can be helium-3. As an example, helium-3 can be used for detection of relatively slow moving thermal neutrons (e.g., neutrons having an energy around about 0.025 eV). Helium-3 can be present in a mount sufficient to provide a partial pressure within the range from about 1.5 Psia to about 150 Psia. The minimum partial pressure represents an estimate of the minimum amount of helium-3 required for viable operation of the proportional counter **100**. In contrast, above the maximum partial pressure, the increase in sensitivity drops dramatically for a given increase in helium-3 partial pressure. Thus, it is economically undesirable to fill above this maximum partial pressure.

In other embodiments, the neutron sensitive gas can be at least one of helium-4 (^4He), hydrogen H_2, or uranium hexafluoride UF_6. As an example, helium-4 and hydrogen can be used for detection of fast neutrons (e.g., neutrons having an energy within the range from about 1 MeV to about 20 MeV), where helium-3 is not suitable. Uranium hexafluoride can be used for detection of either thermal neutrons or fast neutrons. In further embodiments, the at least one neutron sensitive gas has a thermal neutron absorption cross-section within the range from about 100 barns to about 5600 barns. In additional embodiments, the at least one neutron sensitive gas has a fast neutron total cross-section with the range from about 1 barns to about 8 barns.

The gas mixture **110** can further include one or more stopping gases. The stopping gas is different from the quench gas **114**. Notably, the quench gas **114** is configured to terminate the pulse, while the stopping gas has an ionization potential that allows for gas multiplication of the primary ions. Thus, in this context, stopping refers to the ability of the stopping gas to reduce the mean free path of primary ions (e.g., either or both of the ion pair **120**) within the gas mixture **110**. Examples of such stopping gases can include, but are not limited to, at least one of argon (Ar), krypton (Kr), or xenon (Xe).

In further embodiments, a method **200** for preparing a proportional counter is provided. FIG. **2** is a flow diagram illustrating one exemplary embodiment of the method **200**. As shown, the method **200** can include operations **202**-**204**. However, it can be appreciated that, in alternative embodiments, the method can include greater or fewer operations than illustrated in FIG. **2** and the operations can be performed in a different order than illustrated in FIG. **2**.

In operation **202**, a proportional counter is provided. In an embodiment, the proportional counter can be the proportional counter **100** of FIG. **1**, including the chamber **102** having an anode and a cathode.

In operation **204**, the chamber **102** can be filled with a gas mixture **110**. The gas mixture **110** can include the fill gas **112** (e.g., at least one neutron sensitive gas) and the quench gas **114** (e.g., BF_3). Examples of the at least one neutron sensitive gas can include, but are not limited to, He-3, He-4, H_2, or UF_6). In an embodiment, the partial pressure of the fill gas **112** within the gas mixture **110** can be provided within the range from about 1.5 Psia to about 150 Psia. In a further embodiment, the partial pressure of BF_3 within the gas mixture **110** can be within the range from about 0.002 Psia to about 3.9 Psia.

In a further embodiment, the at least one neutron sensitive gas can be configured to detect thermal neutrons, fast neutrons, and/or combinations thereof. As an example, in one example, the at least one neutron sensitive gas can have a thermal neutron absorption cross-section within the range from about 100 to 5600 barns. As another example, the at least one neutron sensitive gas can have a fast neutron total cross-section within the range from about 1 to 8 barns.

In embodiments, the gas mixture **110** can also include at least one stopping gas. The at least one stopping gas can be configured to reduce a mean free path of primary ions within the gas mixture. The primary ions can be electrons or molecules of the fill gas **112** ionized by the incident radiation **116**. Examples of the stopping gas can include, but are not limited to, Ar, Kr, or Xe.

Exemplary technical effects of the methods, systems, and devices described herein include, by way of non-limiting example a proportional counter including a novel gas mixture including a neutron sensitive gas and BF_3 as a quench gas. In one aspect, the gas mixture provides improved sensitivity for detection of neutrons, as compared to gas mixtures without BF_3. In another aspect, decomposition products of the BF_3 quench gas are capable of recombining, providing the quench gas with effectively indefinite life.

Certain exemplary embodiments have been described to provide an overall understanding of the principles of the structure, function, manufacture, and use of the systems, devices, and methods disclosed herein. One or more examples of these embodiments have been illustrated in the accompanying drawings. Those skilled in the art will understand that the systems, devices, and methods specifically described herein and illustrated in the accompanying drawings are non-limiting exemplary embodiments and that the scope of the present invention is defined solely by the claims. The features illustrated or described in connection with one exemplary embodiment may be combined with the features of other embodiments. Such modifications and variations are intended to be included within the scope of the present disclosure. Further, in the present disclosure, like-named components of the embodiments generally have similar features, and thus within a particular embodiment each feature of each like-named component is not necessarily fully elaborated upon.

Approximating language, as used herein throughout the specification and claims, may be applied to modify any quantitative representation that could permissibly vary without resulting in a change in the basic function to which it is related. "Approximately," "substantially, or "about" can include numbers that fall within a range of 1%, or in some embodiments within a range of 5% of a number, or in some embodiments within a range of 10% of a number in either direction (greater than or less than the number) unless otherwise stated or otherwise evident from the context (except where such number would impermissibly exceed 100% of a possible value). Accordingly, a value modified by a term or terms, such as "about," "approximately," and "substantially," are not to be limited to the precise value specified. In at least some instances, the approximating language may correspond to the precision of an instrument for measuring the value. Here and throughout the specification and claims, range limitations may be combined and/or interchanged, such ranges are identified and include all the sub-ranges contained therein unless context or language indicates otherwise.

In the descriptions herein and in the claims, phrases such as "at least one of" or "one or more of" may occur followed by a conjunctive list of elements or features. The term "and/or" may also occur in a list of two or more elements or features. Unless otherwise implicitly or explicitly contradicted by the context in which it is used, such a phrase is intended to mean any of the listed elements or features individually or any of the recited elements or features in combination with any of the other recited elements or features. For example, the phrases "at least one of A and B;" "one or more of A and B;" and "A and/or B" are each intended to mean "A alone, B alone, or A and B together." A similar interpretation is also intended for lists including three or more items. For example, the phrases "at least one of A, B, and C;" "one or more of A, B, and C;" and "A, B, and/or C" are each intended to mean "A alone, B alone, C alone, A and B together, A and C together, B and C together, or A and B and C together." In addition, use of the term "based on," above and in the claims is intended to mean, "based at least in part on," such that an unrecited feature or element is also permissible.

One skilled in the art will appreciate further features and advantages of the invention based on the above-described embodiments. Accordingly, the present application is not to be limited by what has been particularly shown and described, except as indicated by the appended claims. All publications and references cited herein are expressly incorporated by reference in their entirety.

The invention claimed is:

1. A neutron proportional counter, comprising:

a chamber comprising an anode and a cathode; and

a gas mixture contained within the chamber, the gas mixture comprising:

at least one neutron sensitive fill gas; and

a quench gas comprising BF_3.

2. The proportional counter of claim **1**, wherein the fill gas has a thermal neutron absorption cross-section within the range from about 100 to 5600 barns.

3. The proportional counter of claim **1**, wherein the fill gas has a fast neutron total cross-section within the range from about 1 to 8 barns.

4. The proportional counter of claim **1**, wherein the fill gas comprises He-3.

5. The proportional counter of claim **4**, wherein He-3 is present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 1.5 Psia to about 150 Psia.

6. The proportional counter of claim **1**, wherein the fill gas comprises at least one of He-4, H_2, or UF_6.

7. The proportional counter of claim **1**, wherein the gas mixture further comprises at least one stopping gas configured to reduce the mean free path of primary ions within the gas mixture.

8. The proportional counter of claim **7**, wherein the stopping gas comprises at least one of Ar, Kr, or Xe.

9. The proportional counter of claim **1**, wherein BF_3 is present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 0.002 Psia to about 3.9 Psia.

10. A method of preparing neutron proportional counter, comprising:

providing a neutron proportional counter comprising a chamber comprising an anode and a cathode; and

filling the chamber with a gas mixture comprising at least one neutron sensitive fill gas and a quench gas comprising BF_3.

11. The method of claim **10**, wherein the fill gas has a thermal neutron absorption cross-section within the range from about 100 to 5600 barns.

12. The method of claim **11**, wherein the fill gas has a fast neutron total cross-section within the range from about 1 to 8 barns.

13. The method of claim **11**, wherein the fill gas comprises He-3.

14. The method of claim **13**, wherein He-3 is present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 1.5 Psia to about 150 Psia.

15. The method of claim **11**, wherein the fill gas comprises at least one of He-4, H_2, or UF_6.

16. The method of claim **11**, wherein the gas mixture further comprises at least one stopping gas configured to reduce the mean free path of primary ions within the gas mixture.

17. The method of claim **16**, wherein the stopping gas comprises at least one of Ar, Kr, or Xe.

18. The method of claim **11**, wherein BF_3 is present in the gas mixture in an amount sufficient to provide a partial pressure within the range from about 0.002 Psia to about 3.9 Psia.

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